Laser-Stimulated Adspecies Interaction
with a Semiconductor Surface

by

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The surface electronic charge distributions of a one-dimensional semiconductor are compared for the ground state and the laser-excited surface states. A charged adspecies interaction potential with these excited surface states is examined for the case of silicon. The use of a laser to enhance desorption or adsorption by this process is discussed.

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## Abstract

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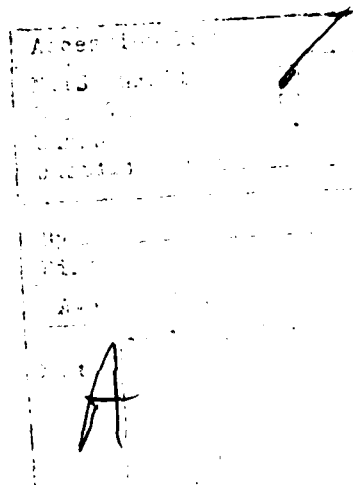
# 1. Introduction

Laser-induced photochemistry with adspecies is a growing
area of interest, with much attention being given to desorption
processes. For example, Djidjoev and co-workers[1] have reported
that the desorption of hydroxyl groups from the surface of silica
can be greatly enhanced in the presence of an infrared laser.
Chuang[2] has also presented experimental evidence that the reaction
of an adspecies with a silicon surface can be stimulated by laser
radiation.

For the theoretical descriptions of these processes, a
statistical analysis has been given by Lin et al.,[3] while Jedrzejck
and his associates[4] have used a simple one-dimensional model. Both
of these theoretical studies, and the experimental works, have
relied on the use of the laser to excite the vibrational modes of
the system.

On the other hand, photo-induced surface reactions can occur
through electronic excitation. Such effects have been demonstrated
by synchrotron radiation studies[5] on metal surfaces. Desorption of
an adspecies is produced by a shift in electronic charge in the
surface region and the resultant Coulombic repulsion.[6]

For a semiconductor with a surface, the electronic energies
consist of various surface states, in addition to the bulk valence
and conduction bands.[7] These surface states, with charge localized
in the surface region, can be populated by using a laser to excite
electrons from the bulk electronic bands.[8] As in the case of
synchrotron radiation, such surface charge could induce a strong

Coulombic interaction with an adspecies.

In the next section, we calculate the electronic density profile of a model one-dimensional semiconductor, after which the effect of surface state excitation on this curve is examined. The changes in the interaction potential of a charged adspecies with the surface in various excited states is then calculated and compared. Finally, limitations and possible extensions of our model and its application to laser-induced surface processes are discussed.

## 2. Electronic Density Profile

In our model, we have a one-dimensional linear chain of atoms of length L and with lattice constant a, which is confined along the z-axis to the region $z < a/2$. Such a model has been shown to produce valence, conduction and surface bands.[7,8] In the ground state, the valence band is completely occupied while the surface and conduction bands are empty.

To obtain the total electron density, $n(z)$, we must sum the individual densities of all occupied states:

$$n(z) = \frac{8L}{(2\pi)^3} \int d^3k |\psi_k(z)|^2 \qquad (1)$$

where the wave function, $\psi_k(z)$, with crystal momentum k was previously determined.[7,8] If we now use cylindrical coordinates, this can be readily reduced to

$$n(z) = \frac{2L}{\pi^2} \int_0^{k_F} dk [E_F - E(k)] |\psi_k(z)|^2 \qquad (2)$$

where $E_F$ is the Fermi energy of crystal momentum $k_F$.

Because of its importance as a semiconductor, we have used parameters[9],[10] typical of silicon to numerically evaluate the density profile, and our results are shown by the solid line in Figure 1. The oscillations of the charge as one goes into the bulk of the crystal and the exponential tail are typical of such density profiles. These results are for the ground electronic state. The effect on this profile of promoting electronic charge by laser action into the surface states is considered below.

### 3. Charge Density of the Excited System

If the system is now exposed to a laser with an energy of about half the band gap, it is possible to populate surface states of our crystal.[8] The wave function of these surface states is of the form

$$\psi_\kappa = C_s e^{-\kappa(z-\frac{a}{2})} \sin[\frac{g}{2}(z-\frac{a}{2})+\theta_\kappa] \tag{3a}$$

for $z < \frac{a}{2}$ and

$$\psi_\kappa = C_s \sin\theta_\kappa e^{-q(z-\frac{a}{2})} \tag{3b}$$

for $z > \frac{a}{2}$, with the energy given by

$$E(\kappa) = \frac{1}{2}[(\frac{g}{2})^2 - \kappa^2 \pm (4V^2 - \kappa^2 g^2)^{1/2}] \tag{4}$$

where the normalization constant, $C_s$, the exponential factor, $q$, and the phase factor, $\theta_\kappa$, have previously been determined.[7],[8] The parameter $\kappa$ in the above expressions is the imaginary part of the crystal momentum that occurs in surface states. $V$, the $g$ component of the Fourier transform of the effective potential where $g = 2\pi/a$ is the reciprocal lattice constant, is assumed to be positive.

Using Equation (1) we find the new charge density to be

$$n(z) = n_0(z) - |\psi_{\bar{\kappa}}(z)|^2 + |\psi_\kappa(z)|^2, \tag{5}$$

where $n_0(z)$ refers to the charge density of the unexcited system given by Equation (2) and the transition proceeds from bulk state k to surface state $\kappa$. As previously pointed out,[8] the excitation conserves the crystal momentum. Therefore, bulk states are excited at the band edge with $k = \frac{g}{2}$:

$$\psi_{\frac{g}{2}}(z) = \frac{2}{L^{1/2}} \sin\left[\frac{g}{2}\left(z - \frac{a}{2}\right) + \theta_{\frac{g}{2}}\right]. \tag{6}$$

However, since the charge associated with this state goes as $1/L$, for a very large system we would in effect be taking only an infinitesimal amount of charge from everywhere in the semiconductor to populate the surface state. Consequently, for a large system, the new charge density can be written

$$n(z) = n_0(z) + |\psi_\kappa(z)|^2. \tag{7}$$

Using Equation (3) in Equation (7) along with the results for the ground state, we have obtained the density of the semiconductor with the $\kappa = -.5(2V/g)$ lower branch surface state excited. The results are depicted in Figure 1 by the dashed line. As can be seen by the plot, this excited state produces an electronic charge in the surface region that is twice as great as the bulk average. If the surface states are excited closer to the branch point near the gap center, $\kappa = -2V/g$, the charge concentration in the first few layers of the surface will increase up to about thrice the average density. The interaction of an adspecies with this charged surface will now be examined.

## 4. Adspecies-Surface Interaction

If there is a charged adspecies above our surface, the classical interaction between this ion and our semiconductor can be written as

$$U(z_I) = - \int n(z) v(r) d\vec{r} \qquad (8)$$

with

$$r = [x^2 + y^2 + (z - z_I)^2]^{1/2}, \qquad (9)$$

where $v(r)$ is the electron-ion potential of the adspecies at $z_I$. Let us assume that $v(r)$ is Coulombic in nature with Thomas-Fermi screening:[11]

$$v(r) = \frac{Ze^{-\lambda r}}{r} \qquad (10)$$

where $\lambda$ is the screening parameter,

$$\lambda^2 = \frac{6\pi\bar{n}}{E_F}, \qquad (11)$$

and $Z$ is the charge or the adspecies. A more appropriate screening may well be similar to those developed for finite metals,[12] but such a calculation is beyond the scope of this paper. Since the Thomas-Fermi screening parameter depends on the average electron density, $\bar{n}$, we would expect this parameter to be less in the surface region. Consequently, the use of a screening parameter based on the bulk density would underestimate the actual interaction.

Bearing this limitation in mind, Equations (8) and (10) can be combined and readily simplified to give the interaction

$$U(z_I) = - \frac{2\pi Z}{\lambda} \int_{-\infty}^{\infty} n(z) e^{-\lambda|z - z_I|} dz. \qquad (12)$$

Inserting Equation (7) into this expression, we obtain

$$U(z_I) = - \frac{2\pi Z}{\lambda} \int_{-\infty}^{\infty} n_0(z) e^{-\lambda|z - z_I| - S} U(z_I) dz, \qquad (13)$$

where the change in potential induced by the excited surface states is given by

$$S_U(z_I) = \frac{2\pi Z}{\lambda} \int_{-\infty}^{\infty} |\psi_\kappa|^2 e^{-\lambda|z-z_I|} dz. \tag{14}$$

Inserting the expression for the surface wave function, we get

$$S_U(z_I) = \frac{2\pi Z C_S^2}{\lambda} \{ \int_{-\infty}^{a/2} \sin^2[\frac{g}{2}(z-\frac{a}{2})+\theta_\kappa] e^{-2\kappa(z-\frac{a}{2})-\lambda|z-z_I|} dz$$

$$+ \sin^2\theta_\kappa \int_{a/2}^{\infty} e^{-2q(z-\frac{a}{2})-\lambda|z-z_I|} dz \}. \tag{15}$$

After much algebra, these integrals yield an interaction potential of the form

$$\frac{S_U(z_I)}{z} = e^{-\lambda z_I} A(\kappa) - e^{-2q z_I} B(\kappa), \tag{16}$$

where

$$A(\kappa) = \frac{C_S^2 \pi \sin^2\theta_\kappa}{\lambda} [\frac{1}{(q-\frac{\lambda}{2})} - \frac{1}{2\kappa-\lambda} + \frac{(2\kappa-\lambda)\cos 2\theta_\kappa - g\sin 2\theta_\kappa}{(2\kappa-\lambda)^2+g^2}] e^{\lambda a/2} \tag{17}$$

and

$$B(\kappa) = \frac{C_S^2 4\pi \sin^2\theta_\kappa}{(2q)^2-\lambda^2} e^{qa}. \tag{18}$$

Equation (16) has been evaluated for a number of surface states, and the results are plotted in Figure 2. The curves clearly show that as one moves to larger $|\kappa|$ (energies near the gap center), both the intensity and the range of the surface charge interaction increase, as would be expected from the density calculation. All curves, however, show an appreciable contribution to the potential produced by the surface states with $|\kappa| > 0.1(2V/g)$.

A better comparison of the surface charge interaction among the various surface states is given in Figure 3, where we have plotted the change in potential at $z_I = a$ for all the surface states.

The upper branch states are at a higher energy [positive sign in Equation (4)] than the lower branch states. Therefore, the exponential tail and, subsequently, the interaction is slightly greater.

## 5. Discussion

Use of a laser to localize electronic charge in the surface region of a semiconductor can produce an appreciable effect on an adspecies-surface interaction. If the adspecies is negatively charged, desorption can be induced; if positively charged, the possibility of adsorption is enhanced. In a more realistic model with both occupied and empty surface states, the laser excitation of holes as well as electrons could be used to selectively enhance adsorption or desorption for the same charged species.

Since the concentration of charge is so great in the surface region, as shown in Figure 1, one would expect the effective interaction length to be greater than that indicated by Figure 2. The exponential decay of our potential is probably an artifact of the assumed Thomas-Fermi screening. An improved interaction potential would necessitate addressing the dielectric screening problem in more detail.[12]

Another consideration is the fact that we did not look at the equilibrium concentration of the surface state - only one surface state was assumed to be populated. On the other hand, several partially occupied surface states may well have as much as or more surface charge than a single occupied surface state.

Of course, the major limitation of our model is its one-dimensionality. The three-dimensional interaction potential may be quite complex depending not only on the distance from the surface

but also on the position of the adspecies with respect to the plane of the surface. Also, many of the features of a crystal cannot be adequately treated in a one-dimensional model.

Nonetheless, our contention that lasers used to control surface charge density can lead to enhanced surface processes has been substantiated. The effects on this process of higher dimensions, phonons, and the dynamics of the adspecies and surface are the subject of continuing research.
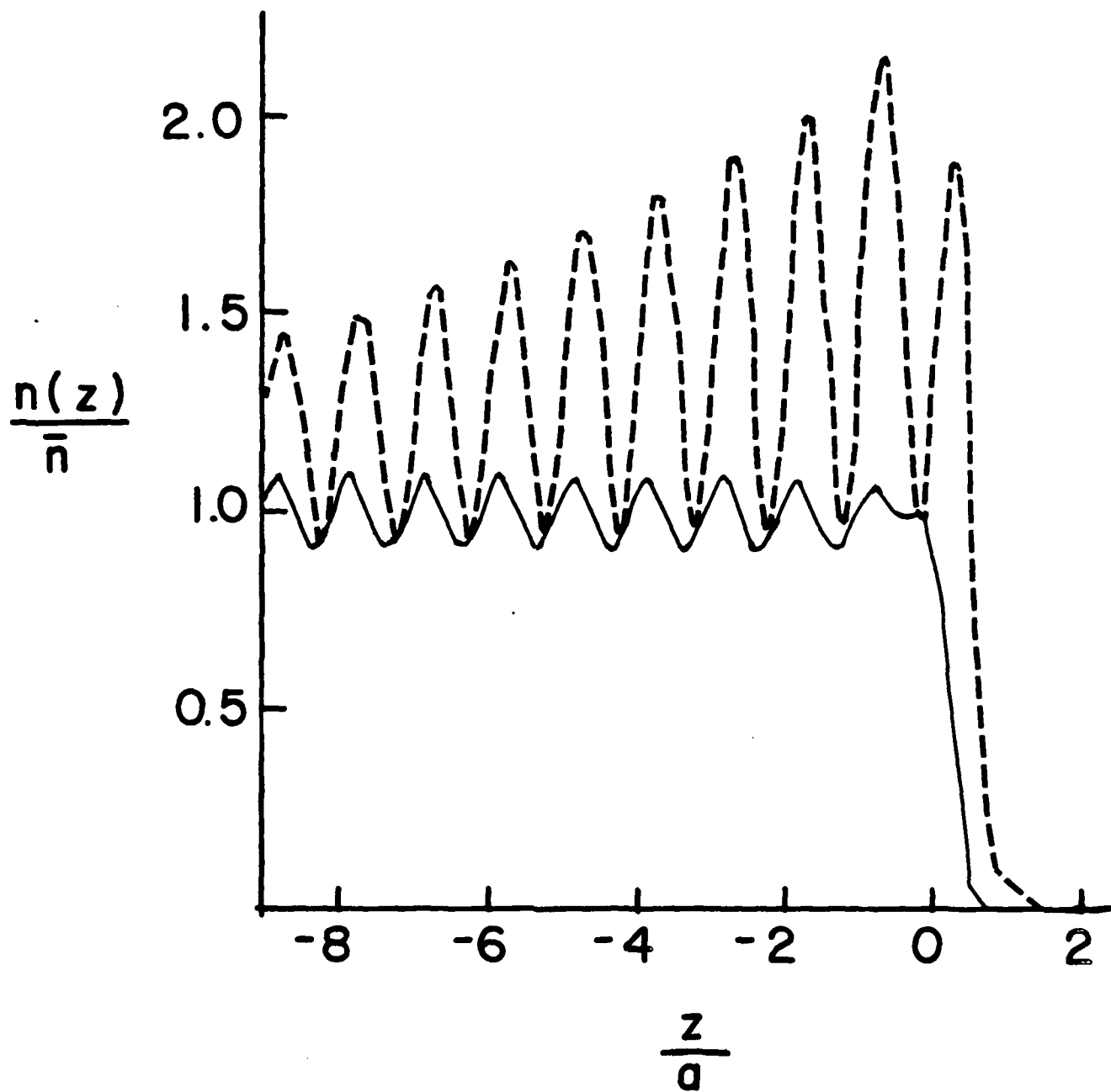
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Figure 1.  Electron density distribution at the surface.  The solid line represents the ground electronic state, and the dashed line represents the system with the excited surface state $\kappa = -.5$ $(2V/g)$ in the lower branch.

Figure 2.  The magnitude of the surface interaction potential (in millihartrees) at various distances from the surface.  The solid line represents the system with excited state $\kappa = -(2V/g)$; the dashed line, $\kappa = -.5(2V/g)$; and the dotted line, $\kappa = -.1(2V/g)$, all in the lower energy branch.

Figure 3.  The magnitude of the surface interaction potential (in millihartrees) at a distance $z_I = a$ for the system with various excited surface states.  The solid line represents surface states in the lower energy branch; the dashed line, the upper energy branch.
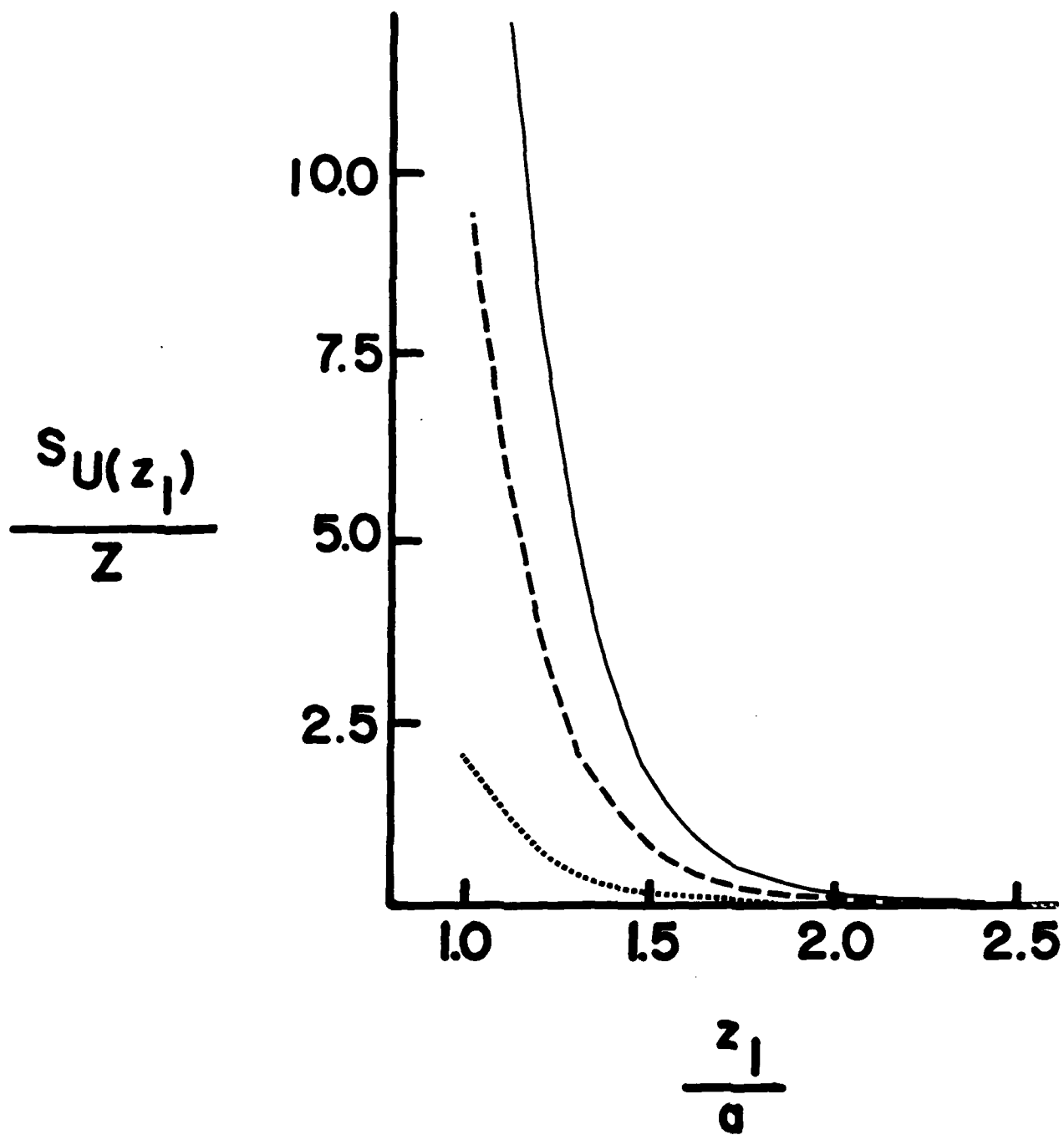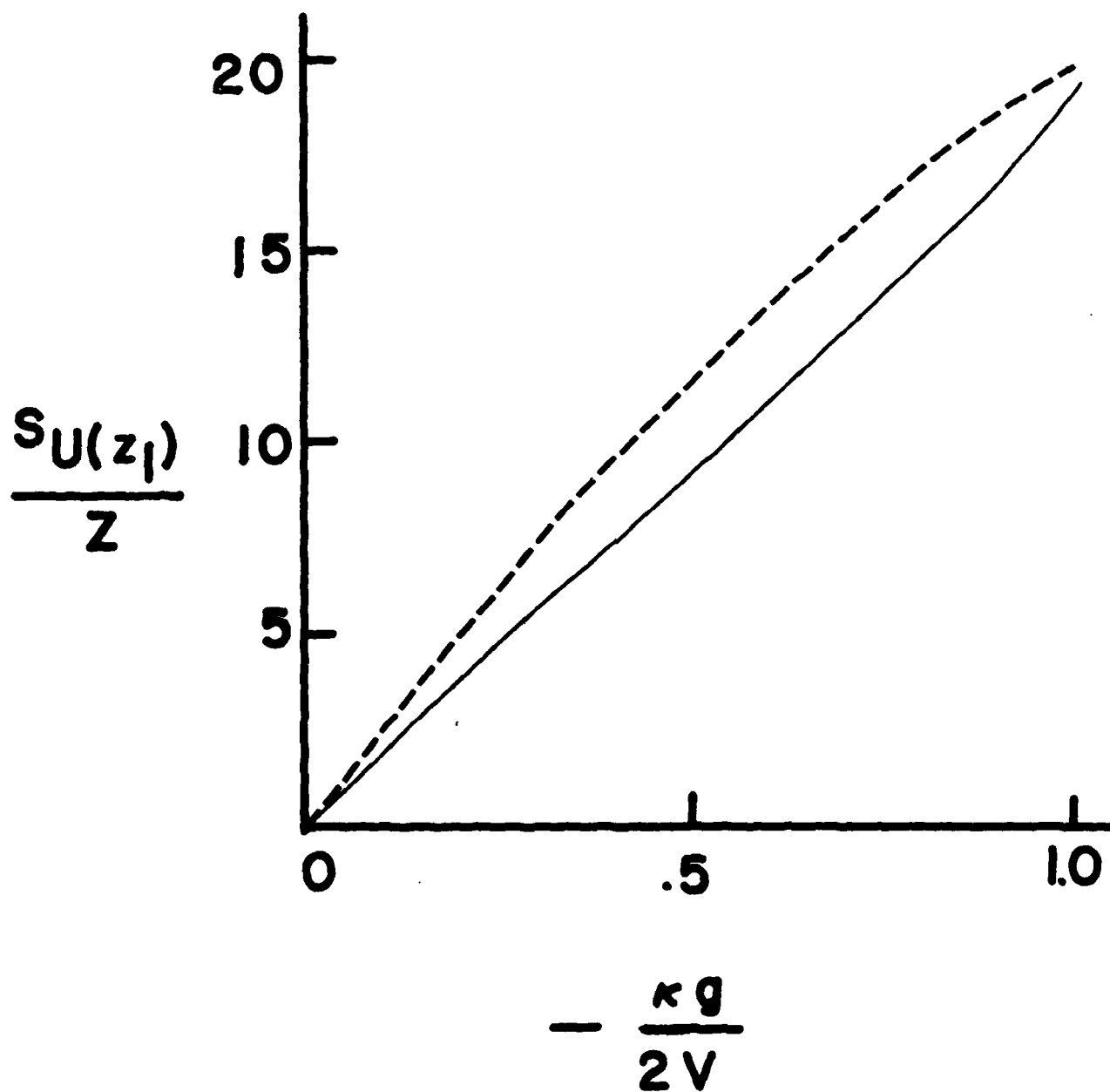
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